Final Report
Grant No. N00014-91-J-1850
June 1, 1991 - December 31, 1994

APPLICATION OF NONLINEAR SIGNAL PROCESS TECHNIQUES

Submitted to:

Office of Naval Research
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Arlington, VA 22217-5660

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Report No. UVA/525456/CHE95/101
May 1995

DEPARTMENT OF CHEMICAL ENGINEERING

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UNIVERSITY OF VIRGINIA
School of Engineering and Applied Science

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REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY *(Leave Blank)*

2. REPORT DATE
May 1995

3. REPORT TYPE AND DATES COVERED
6/1/91 - 12/31/94 Final

4. TITLE AND SUBTITLE
Application of Nonlinear Signal Process Techniques

5. FUNDING NUMBERS
N00014-91-J-1850

6. AUTHOR(S)
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7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES)
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8. PERFORMING ORGANIZATION REPORT NUMBER
UVA/525456/CHE95/101

9. SPONSORING MONITORING AGENCY NAME(S) AND ADDRESS(ES)
Office of Naval Research
800 N. Quincy Street
Arlington, VA 22217-5660

10. SPONSORING/MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION/AVAILABILITY STATEMENT

12b. DISTRIBUTION CODE

13. ABSTRACT *(Maximum 200 words)*

See Text of Report

Accesion For		
NTIS CRA&I	☒	
DTIC TAB	☐	
Unannounced	☐	
Justification		
By		
Distribution /		
Availability Codes		
Dist	Avail and / or Special	
A-1		

14. SUBJECT TERMS

15. NUMBER OF PAGES
26

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
Unclassified	Unclassified	Unclassified	UL

Standard Form 298 (Rev 2-89)

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12 April, 1995.

OFFICE OF NAVAL RESEARCH

FINAL REPORT

- **Contract/Grant Number:** N00014-91-J-1850
- **Contract/Grant Title:** Application of Nonlinear Signal Processing Techniques to Chemical and Transport Processes
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1.1 RESEARCH ACCOMPLISHMENTS:

During the course of the grant we have carried out -as outlined in the original proposal- computational and experimental studies of nonlinear reaction and transport processes, collected time series and image series from these processes, and we also developed, implemented, tested and applied algorithms for nonlinear signal processing and system identification on these time series. We also

- developed (in collaboration with the T-13 group at Los Alamos National Laboratory) an integrated computational environment for the generation of such nonlinear models from time series, based on artificial neural networks;

- studied both computationally and experimentally the dynamics of adaptive control algorithms;
- collaborated with the groups of Dr. Carlos Garcia at Shell and of Dr. Barry Tarmy at Exxon;
- motivated by the model reduction and image processing work, constructed a new class of microstructured and composite catalytic materials.

Detailed research accomplishments (both planned in the original proposal and new directions that arose as parts of the research over the years) are described below, with references to the attached publication list. We will include a short description of a number of ongoing projects which have been motivated by this work, and which, when completed, will still acknowledge this grant.

1.1.1 Time Series Analysis Collection and Analysis

The bulk of the research consisted of the application of artificial neural network (ANN) based techniques to the processing of experimental (and occasionally computational) time series. The experimental time series for metal electrodissolution were obtained at our laboratory at the University of Virginia (Cu electrodissolution in phosphoric acid, metastable pitting of Al and Al-Cu alloys in halide solutions); experimental time series from thermal convection were obtained in collaboration with Dr. R. E. Ecke, of the MST-10 division at Los Alamos National Laboratory.

Our original collaboration, on which the proposal was based, appeared in *Chem. Eng. Science* in 1990; from the ANN point of view, its originality consisted of the incorporation of an additional input neuron to model the effect of an operating parameter on the system dynamics. This allowed the modeling of an oscillatory and a period-doubling instability for Cu electrodissolution in phosphoric acid solutions, and we are happy to report that the paper has been reprinted in a book entitled "Artificial Neural Networks, Forecasting Time Series," V. Rao Vemuri and Robert D. Rogers, eds., IEEE Computer Society Press, Washington (1994) (ISBN 0-8186-5120-2).

This work continued and resulted in our *Chem. Eng. Communications* publication in 1992. Here, Cu electrodissolution and a sequence of

period-doublings leading to chaotic dynamics was studied carefully and in detail. Furthermore, the paper contained two innovations in our neural network research. The first (which was part of the title of the paper) was the development and implementation of a new class of neural network architectures -templated on numerical integrators- which could yield continuous-time models as opposed to discrete-time ones. This development allowed for the qualitatively correct interpretation of instabilities and bifurcations, which "traditional", discrete-time ANN or other models cannot capture. This work has received some recognition, and recently we made our codes available to Professor T. McAvoy, of the University of Maryland. The second development was the use of so-called "nonlinear principal components" for the preprocessing of the time series; the results of this preprocessing are then fed to the continuous-time ANN algorithms.

Our 1994 *Physica D* paper on processing time series from the quasiperiodic regime of Rayleigh-Benard convection did not contain similar novelties in neural network architectures; nevertheless, it was the first to attempt to explain truly complex sequences of *global* bifurcations for Poincaré maps in a very rich dynamical regime; this paper was initially a Los Alamos report, was made electronically available at Los Alamos, and we received a large number of copy requests within a few days of its appearance.

After successfully completing the analysis of the thermal convection data, our efforts on the development of neural network algorithms turned to the novel class of integrator-based architectures we proposed for continuous-time system identification. While in our *Chem. Eng. Communications* paper we proposed the use of *iterated* neural networks -based on integrators like Runge-Kutta-type algorithms- it became clear that for "stiff" systems one would need ANN architectures based on *implicit* integrators, and these give rise to *recurrent* neural networks. The efficient training of such networks (currently done by essentially "feedforward" algorithms, like the one proposed by Pineda in 1987), is still an open problem, and we have made significant progress in that direction, as discussed also below in the algorithm section. We have made several presentations on this work, including a refereed proceedings paper at the 1993 IEEE NN Conference in San Francisco, a proceedings paper in the 1993 6th SIAM Conference on Parallel Computing and Applications, as well as a proceedings paper in the 1993 American Control Conference in San Francisco.

This work has also naturally branched towards the identification of

gray box models, based on neural network architectures; the first publication along these lines has been a refereed proceedings paper in the 1994 IEEE Workshop on Neural Networks for Signal Processing. We are currently continuing this work towards parallel implementation of these algorithms using PVM on an IBM SP2 parallel computer, a small part of which was financed through this grant. Both of the last directions (recurrent black box ANNs templated on implicit integrators as well as gray-box-type ANNs, also based on implicit integrators) are still amenable to the development of new training algorithms to exploit massively parallel architectures. While Pineda-type algorithms "fit" SIMD machines, some of the more "exact" algorithms we have developed and implemented in scalar architectures and environments like PVM can be much better fitted to MIMD machines, and that is a subject we will continue to do research on.

A final aspect of our ANN research was the realization (motivated by our study of adaptive control systems) that discrete-time neural network models based on time series can be *noninvertible*, *i.e.* a given state can have more than one "preimages" backward in time. This is a pathology, which -as in our work above about discrete-time-modeling shortcomings- must be kept in mind, and tested for, before one trusts the predictive power of discrete-time ANNs. We have published so far a refereed proceedings paper in the 1993 IEEE NN conference proceedings, as well as a paper in the proceedings of the 28th annual IEEE conference on Information Systems and Sciences (1994) on this matter. We are currently completing an archival journal paper on this subject, which we will submit to IEEE Journal on Neural Networks during this academic year.

We were invited to write a chapter on these two issues (discrete- vs. continuous-time ANN models, as well as noninvertibility in ANNs) in an Elsevier book ("Neural Networks for Chemical Engineers", A. Bulsari ed.); we were just notified that the book has been published.

On the study of metastable pitting of aluminum and aluminum alloys an intensive experimental and mathematical study program was initiated at Virginia. It is known that metastable pitting occurs at potentials well below the pitting potential in many alloy-electrolyte systems. An understanding of the metastable pitting process is important not only in its role in the stable pitting process but also independently in miniaturized components such as in VLSI circuits. We have developed an apparatus for obtaining low noise current signals in the nanoamp range. Analysis of the signals (*J. Electrochem.*

Soc. (94)) shows that they can be modeled as a stochastic process with the probability of initiation being dependent on previous events.

Professor Hudson was also invited to give one of the plenary lectures in the 2nd Experimental Chaos Conference, and contributed a review article on "Chaos during heterogeneous chemical reactions" in the Proceedings of that conference.

The chaotic time series from the electrodissolution of copper have also been analyzed by a related global vector field method in cooperation with a group at the Laboratoire d'Energetique des Systèmes et Procédés at Rouen, France. In this work (which just appeared in *J. Phys. Chem.*), it is shown that the attractor obtained from the reconstructed system is topologically equivalent to the attractor obtained directly from the experimental data.

1.1.2 Spatiotemporal Dynamics

One of the main issues of the research we originally proposed was to move beyond scalar time series, and develop / exploit model identification techniques for distributed systems. We had several successes in this direction, and, as we will discuss below, one of the high points of our work (quite unforeseen at the beginning) came from this research direction.

The first success we had came from a collaboration with the group of Professor G. Ertl, at the Fritz Haber Institut of the Max Planck Gesellschaft in Berlin, Germany. This is simply one of the leading surface science / catalysis / electrochemistry groups in the world. In the late 80s they developed an electron microscopy technique called PEEM, or photoemission electron microscopy, which allows the real-time, micrometer resolution observation of reactant adsorbate coverages on metal catalysts in reactions like the CO oxidation on Pt, or NO and CO on Pt etc. This microscopy revealed that catalytic rate oscillations are *not spatially uniform*, and that a bewildering variety of spatiotemporal two-dimensional patterns (spirals, stripes, targets, hexagons, chemical turbulence) happens on the catalyst surface in real time and on a five- to ten micrometer scale. In collaboration with a DFG-postdoctoral Fellow in Princeton (Dr. Katharina Krischer) we analyzed real-time spatiotemporal video PEEM data, and were able to reduce image series (of the order of 60,000 time series) to only *four* relevant time series, based on principal component analysis. Subsequently, we were able to construct an ANN-based model that successfully *predicted* the spatiotem-

poral behavior; this input-output model had four inputs and one time-delay, hence a total of eight degrees of freedom - a surprising, even though not completely unexpected reduction since the data did exhibit spatial coherence. The paper that resulted from this work was published in the *AICHE Journal* in 1993, had excellent reviews, was the first paper in the Journal to contain color pictures, and was excerpted in *Chemical Engineering Progress* in early 1993.

In our invited chapter (mentioned above, which just was published) we were able to further reduce the degrees of freedom in the model using the so-called "Non-Linear Principal Components"; this ended up giving us a probably minimal three-degree of freedom model, which completely predicted the spatiotemporal image series in continuous-time - with just three scalar initial conditions.

A sequence of that work was the analysis (without modeling) of spatiotemporal data from another catalytic reaction, the NO + CO reaction, from the group of Dr. Ronald Imbihl in Prof. Ertl's Laboratory. This work was done mainly by a joint Virginia-Princeton postdoctoral fellow, Dr. Michael Graham, partially supported through this grant, who now is an Assistant Professor of Chemical Engineering at the University of Wisconsin-Madison. The paper from this work is finally in press in *Chaos, Solitons and Fractals* - we were sent the page proofs last month.

An important experimental breakthrough at Prof. Hudson's lab in Virginia, was the discovery of spatiotemporal oscillations in two spatial dimensions in the electrodissolution of iron - a circular electrode, close to the Flade potential, dissolves in a time-dependent manner; the oscillations are associated with the formation and dissolution of a film, which clearly shows spatiotemporal symmetry breaking. The data were collected at Virginia and processed in Princeton, and the publication has appeared in *Physics Letters A* in 1993. A similar study has also been carried out on a ring geometry (*Ind. & Eng. Chem.*, 1995). The spatiotemporal period doubling is again observed; this is followed by another symmetry breaking resulting in patterns on four quadrants of the ring. This work continues in the laboratory of Professor Hudson.

An unexpected development from our catalytic pattern formation work was the idea and the subsequent construction and testing of microstructured heterogeneous catalysts. The motivation was really mathematical: simulations of pattern formation with reaction-diffusion models are done in finite

computational domains, which contain usually a few resolved "features" (spirals, waves, fronts). The actual Berlin experiments were done on catalysts of typical dimension 1cm, while the typical pattern size is 5-10 microns. Obviously, it would make sense, in order to compare experiments with theory, to do experiments in smaller domains, say 20 to 50 microns wide. We thought of using lithography to construct such domains on Pt catalysts, and used titanium as the inert "building material" for "fencing in" finite catalyst domains. This work has led to an amazing variety of spatiotemporal patterns, and has allowed the detection of several new phenomena involving the interactions of patterns with boundaries. We were fortunate to have the original paper from this work published in *Science* in 1994, while the long version of this work is currently in press in *Physics Reports E.* The work started with Dr. Michael Graham, a joint postdoc partially supported through this grant, and continues with Dr. Markus Baer, a DFG Fellow in Princeton. There are several forthcoming papers motivated by and continuing this work (like a recent submitted *Phys. Rev. Letter*), and there is a novel direction for it described in the "other initiatives" section below.

1.1.3 Software Development

Parallel Algorithms for Neural Network Training

At the T-13 group of Los Alamos National Laboratory the two investigators involved in this research project (Dr. Alan Lapedes and Rob Farber) have been developing, since 1986, a Neural Network Compiler system. This effort formed one of the bases of the research proposed and performed under this grant. This Compiler system allows us to specify an arbitrary neural network architecture and an arbitrary training set (in the appropriate format), and constructs an efficient neural network simulation for the specified destination computer. The target architecture for the production runs for nonlinear system identification based on time- and image-series processing for this research was initially the Thinking Machines CM-2 and CM-200 at Los Alamos.

As part of this grant, in addition to the production runs for the various physicochemical and engineering systems we studied, and which have been reported in the publications that resulted from this work, The Los Alamos team was able to make two critical additions to the compiler system. These

changes were crucial in enabling us to study some of our test cases. The first was incorporation of the ability to simulate recurrent neural networks (networks with both feedforward and feedback connections). The second was to add the ability to generate efficient simulations for the Thinking Machines CM-5 line of parallel supercomputers.

Adding these two new paradigms, recurrent neural networks and support for a MIMD (Multiple Instruction Multiple Data) model computers, required significant rethinking and restructuring of the internals of the compiler. Since the compiler attempts to efficiently exploit every feature of the destination architecture, almost every facet of our system from the model generation, dependency analysis, code optimization, loader/linker, pseudocode emulation and destination machine code generation had to be re-thought and modified.

The efficient simulation of recurrent neural networks is still an open question in the neural network literature. There is no definitive method for the training and evaluation of recurrent neural networks without substantial computational effort. Neural networks are "trained" by adjusting parameters within the network architecture so that the network can reconstruct the desired output vectors of the training set given the input vectors with a minimal error. This is generally accomplished by iteratively evaluating and changing the parameters of the neural network according to some optimization procedure (such as Conjugate Gradients). However, the addition of recurrent connections to the neural network architecture requires that for each input vector the network must somehow reach a fixed point before the match with the target output vector can be computed. This adds serious complications, as the iterative procedure necessary for the simple evaluation of the network output may not converge to a fixed point but may instead oscillate or worse diverge. We used the method of Pineda for the implementation of recurrent network architectures in our compiler. An important characteristic of this algorithm is that the network must be evaluated in a feedforward manner a number of times for each input vector. We discovered that our accuracy requirements dictated a high number of feedforward iterations for each input vector. This could in general add up to two orders of magnitude to the runtime growth of our simulations. Our compiler modifications, as well as significant amounts of computer time were therefore necessary to test and use these algorithms.

The compiler gets its speed on a parallel computer by mapping the

computation to the hardware, so that it becomes a purely computational problem with minimal communications overhead. During execution of a feedforward neural network code, which is being trained according to a least mean squares criterion on the CM-5, the only communication required is that of a global summation across each of the output neurons. We perform this mapping ourselves, because current parallel machines have a high cost in communicating arbitrarily between processors. On the CM-5, the overhead for this communication can be as high as 1000 times the cycle time of the computer. Conversely, communicating from one processor to all the other processors generally requires only one cycle. Our simulations exploited both these characteristics of the communications to efficiently map the neural network simulation to the computational hardware, so that each processor spends its time calculating instead of waiting for data communications. The overall compiler paradigm can be seen in the block diagram of Figure 1. The compiler is also intelligent enough to maintain the recurrent variables within the local processor memory so that a recurrent neural network run requires an expensive global summation only after the network has iterated towards a fixed point. Efficiently doing floating point calculations on the CM-5 architecture has the additional complication that all the data be formatted so that it can be accessed and efficiently loaded into the vector pipeline of the local processor vector unit. A large number of calculations can be avoided if the data to be calculated is boolean in nature (i.e. having values of only zero or one). Depending on the network architecture, the compiler can have neurons and connections specified as being locked to a constant value or as "equivalenced". This was crucial in implementing some of the "continuous-time" identification architectures proposed by the Princeton group.

To facilitate interoperability between the Los Alamos and Princeton software tools, we implemented the expression of the neural network prediction as either a pseudo-code suitable for immediate evaluation within the automated framework of the Los Alamos compiler tools, or as either a C or FORTRAN subroutine suitable for easy integration into a variety of software packages including those developed and used at Princeton. Additionally, we implemented several "pruning" heuristics within the training of the compiler to minimize the size and connectivity of the final trained network. We noted two important effects:

- For many problems the network can sustain the loss of many parameters

Figure 1: Block Diagram of Neural Network System

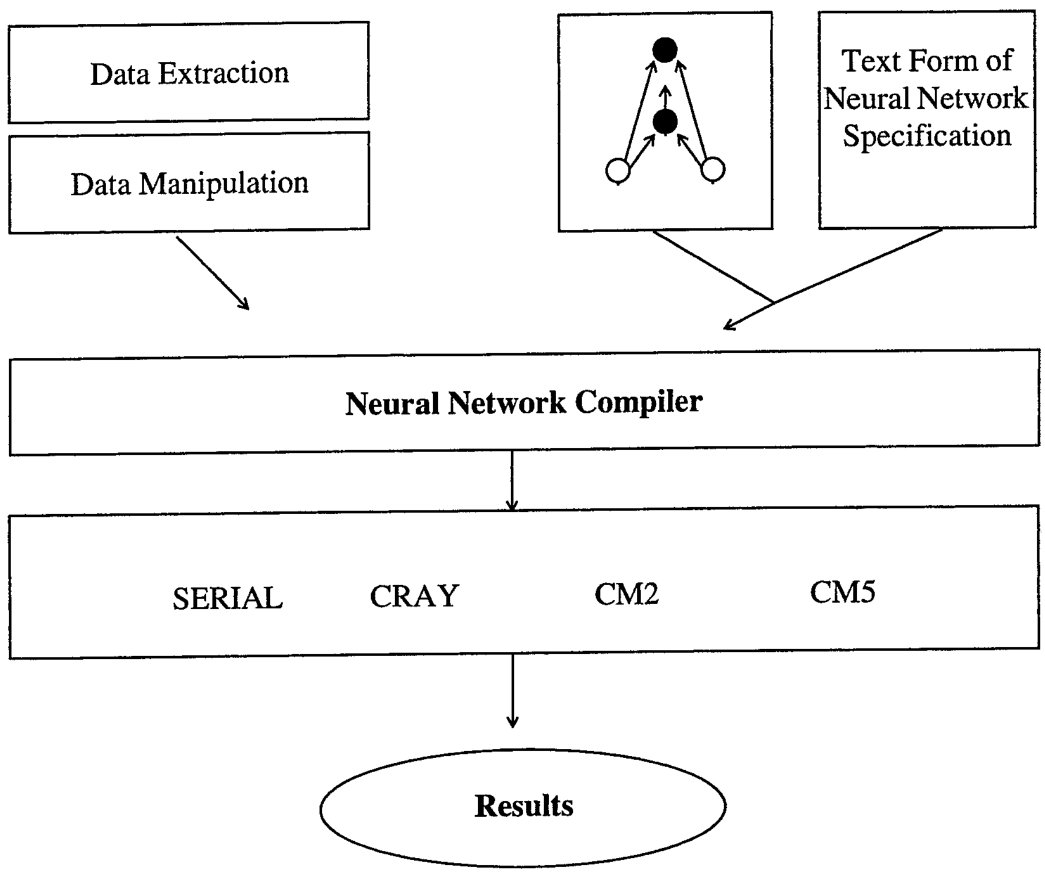
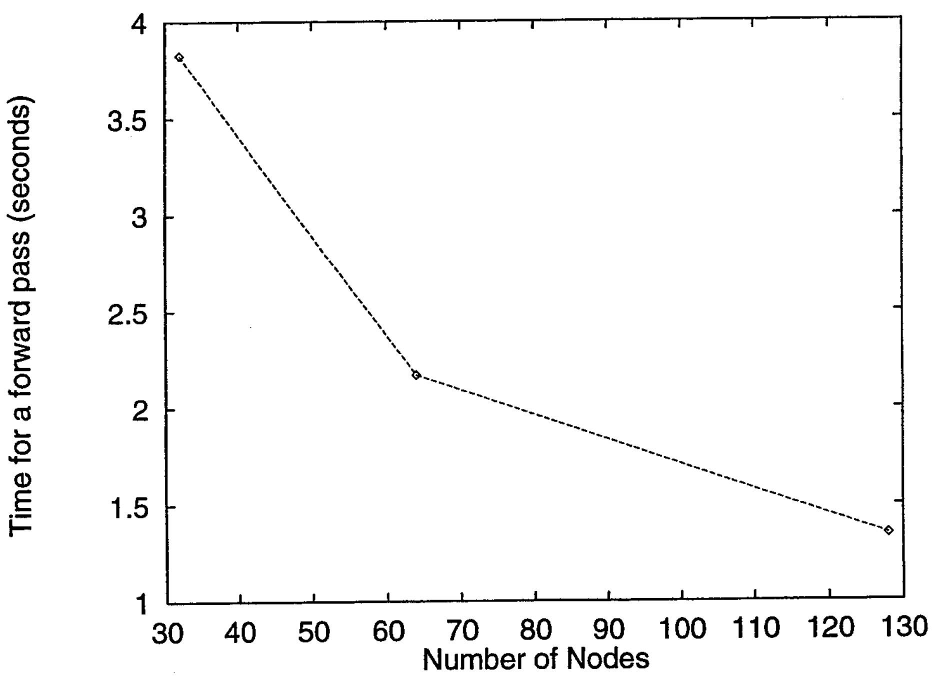


Figure 2: CM-5 timings for a fixed architecture



(neurons and connections) without affecting its generalization properties; and

- The resulting prediction code could gain many multiples in speed increase.

Both effects were helpful in our analysis of the trained networks since they were typically simpler and real time evaluation was significantly faster. Although the compiler is dependent on the computing and communications environment, we have been able to use our code at a number of different institutions for a variety of collaborations on widely varying problems. We are open to collaborations with researchers on problems of interest for which our analytic skills and computational tools can provide a means of solution.

To optimize the training procedure of the computationally efficient simulation generated by our compiler, we use several types of gradient based optimization algorithms. These methods have been shown to be, in general, significantly faster at finding minima than non-gradient based ones. Thus our compiler has the ability to calculate the symbolic derivatives of the specified neural network (recall that the network can be composed of arbitrary "neuron" functions as well as an arbitrary energy function). As long as the functions used are differentiable, the compiler can determine the symbolic derivative of the network and energy function as a whole and express it in efficiently executable code for the destination machine (via the same algorithms which efficiently express the neural network function itself). To the extent that nonlinear optimization is indeed a "black art", we have implemented within the compiler system several optimization routines so we can search for th best-suited to solving our particular problem.

Empirical results indicate that our method of mapping and utilizing the CM-5 architecture is indeed effective. Thinking Machines has acknowledged our implementation of neural networks on the CM-5 as the "most efficient method possible" (Parallel Computing, 14:305-315, 1990). We can see in Figure 2 that for a fixed neural network architecture, the runtime of the forward pass scales well with the number of processors. This of course assumes that there is enough data within the training set to keep the vector pipelines within the CM-5 processors fully loaded. The data for the plot in figure 2 was determined by taking the best time of 50 network evaluations on the 32, 64, and 128 node partitions of the Los Alamos CM-5. Each partition was running in time sharing mode. The neural network architecture was kept

fixed for all the runs. We were unable to run on the 1024 node partition of the Los Alamos CM-5 to provide timings on larger partitions. We have timings for the Naval Research Laboratory 1024 node CM-5 (the use of which was graciously granted to us by ONR through this grant) but we have not included them here as the NRL machine uses different speed processors and hence would not provide an equivalent hardware platform.

Simulation and Stability/Bifurcation Software

The output of the above training procedure is, in subroutine form, a discrete-time neural network state-space model. This subroutine (along with an additional subroutine that calculates first derivatives of the model with respect to its variables and parameters) is then input in a general purpose simulation/ stability analysis/ visualization package that we have been developing over many years in Princeton.

The package is called SCIGMA, and it can be used to study discrete dynamical systems (like the maps produced by traditional discrete-time ANNs) as well as continuous dynamical systems (ODEs, produced by our novel continuous-time ANNs) and also discretized PDEs.

We have made this package available via FTP from Princeton to many researchers in the US and abroad, and we also have used it in teaching graduate and undergraduate classes in Princeton (such as Differential Equations and Introduction to Nonlinear Dynamics).

The package is capable of interactively

- dynamically simulate maps, ODEs and discretized PDEs (currently the maximum system dimension is around 100 for real-time purposes);
- perform real-time three-dimensional interactive visualization of the dynamics on selected projection of the phase space;
- allow the additional real-time visualization of spatiotemporal patterns by reconstructing 1-D (and even 2-D) PDE solutions in physical space and playing them as movies in time;
- locate and analyze the stability of fixed and periodic points for maps;
- locate and analyze the stability of steady states of ODEs and (discretized) steady states of PDEs;

- locate and analyze the stability of limit cycles of ODEs and PDEs finding them and their eigenvalues (Floquet multipliers) as fixed points of interactively determined Poincaré maps;

- approximate 1-dimensional invariant manifolds for fixed or periodic points of maps;

- approximate 1-dimensional invariant manifolds for steady states of ODEs;

- approximate two-dimensional invariant manifolds for steady states of ODEs;

- approximate two-dimensional invariant manifolds for limit cycles of ODEs;

- record and replay a session, automatically scanning a parameter range;

- generate video movies of a session with annotations;

- for the case of discrete neural networks that are noninvertible we had to add additional capabilities, like computing the (several possible) inverse maps and keeping track of the exploding number of successive preimages backward in time, as well as the calculation of the so-called critical curves; this is an experimental version of the code, and this part has not been made publicly available.

In addition, we have built an interactive interface, running on SGI machines using the GL library, for the general purpose bifurcation package AUTO, with the approval of the package's author, Professor E. Doedel of Concordia University in Montreal, Canada.

Our "Interactive AUTO" allows the real-time visualization of continuation and stability calculations for

- one-parameter continuation of steady states of ODEs;

- one-parameter continuation of limit cycles for ODEs;

- one-parameter continuation of fixed and periodic points for maps;

- detection and two-parameter continuation of turning points;

- detection and two-parameter continuation of Hopf points;
- detection and two-parameter continuation of period-doubling points etc.

The original program itself had these *scientific computing* capabilities but in a batch processing environment. Our contribution was to make that real-time interactive *with* real time data visualization, and we were also able to "tailor" the FORTRAN or C output of our neural network training algorithms to the subroutines necessary for linking into this program. This procedure is not completely automated (some hand-editing has to be done before compilation), but could in principle easily become so, especially by running the training output through a symbolic manipulator like Mathematica.

It is worth adding that we are currently "experimentally" working on modifications of these packages for noninvertible systems, for 2-D PDEs, for getting the two packages to completely communicate etc. It is also worth adding that these packages can be used with, in principle, *any* neural network model; they study dynamics of the model one puts in; it is not necessary that this model "come" from one of our own neural network training algorithms.

Finally, we should also mention that in the process of this work we had to develop (with the assistance of the Interactive Computer Graphics Laboratory, in Princeton) a number of programs for the digital access, processing and real time visualization of video data, using optical disks and SGI computers. At the time that we were performing the research, no standards for this type of work were available; this seems to be changing rapidly.

1.1.4 Other Developments

Adaptive control

One of the main motivations of our research on nonlinear signal processing and model identification was the eventual use of these models for control purposes. Our original connection with the Control group at Shell Research and Engineering was Dr. Melinda Golden, who was working on adaptive control problems. We started research on Model Reference Adaptive Control both theoretically and computationally before this proposal, and then as part

of this proposal we studied the effect of plant/model mismatch in adaptive control. This resulted in one publication co-authored with Dr. Golden (Proceedings of a NATO summer school in 1992) as well as the discovery and subsequent analysis of a new secondary instability in an adaptive control system (*SIAM J. Math. Anal. (1995)*). We have also done experimental work on the adaptive control of a mixing tank, which has been published in the Proceedings of the 1992 ACC. Shell R&E initially supported this research financially, and later they donated to the Princeton group a two-tank adaptive control experiment, which we had previously analyzed theoretically, and which was studied in the Senior Thesis of an undergraduate student in Princeton. As part of this work we also consider our comparison of *a priori* theoretical and semi-empirical methods for model reduction in distributed nonlinear systems (a study of the relation between Approximate Inertial Manifold techniques and the POD, or Karhunen-Loéve expansion, with Dr. Michael Graham, which is currently in press in *Computers and Chemical Engineering* (1995)). We are very much still interested in the use of our ANN-identified models in model based control, and hope to continue research along these lines.

Industrial Time Series

During the first year of this work a graduate student from Princeton partially supported through this grant (Dr.Christos Frouzakis) visited Shell R&E for the summer at Westhollow, Houston, Texas, and worked with Dr. Garcia's group in the construction of ANN-based models of a coal gasification process. The time series were obtained by Shell (from a coal gasification plant; the plant was under control –"controllers not tightly tuned"– and inputs where generated during pseudo-random binary noise –PRNB– testing) and were "shifted" in an unknown (to us) way before we were allowed to work with them. The work led to a "second proposition" by Dr. Frouzakis, entitled "Neural Network Identification of Real Plant Data", and was of practical interest to Shell. Unfortunately, as part of restructuring at Shell, that Control group was radically changed, and though we continue being in contact with them (as evidenced by their donation to us of their adaptive control experiment) the interest has shifted from nonlinear system identification.

We also have discussed with Exxon (in New Jersey) the possibility of obtaining data from trickle and fluidized beds. Exxon did partially support our work through an Exxon Education Foundation grant ($10,000 a year for

the last three years), and we are currently discussing with them the donation of a magnetic fluidized bed, which would allow us to look at pattern formation in such gas-particle flow industrial reactors. We did, however, in preparation for this possibility, perform extensive research on the modeling and pattern (bubble) formation in fluidized beds, and a long and systematic paper on this was just submitted to the *J. Fluid Mech.* in February 1995.

Composite Catalysts

One of the "unforeseen" developments of this research, as we discussed above, was the conception, construction, study and modeling of microstructured catalysts, on which spatiotemporal patterns due to catalytic reactions can be observed. While at the beginning we used micropatterning techniques (such as lithography) to "build inert fences" on catalytic surfaces, we now have gone on to the construction of *composite* catalysts: for example, we can -and do- construct checkerboard patterns of one catalyst (say Palladium) on a single crystal of another catalyst (say Pt) at a few microns scale. This new class of composite catalytic materials have the potential of drastically changing the overall reactivity or selectivity of the catalysts for some reactions. For example, a small circle, a couple of monolayers thick, of one catalyst on another may act as a "pacemaker" for the extended surface by facilitating the adsorption of a key species, which is then transported through surface diffusion to the rest of the surface.

We are of course fortunate that the relevant scales (microns) are at the same time relevant (comparable to surface diffusion length scales), accessible to the available microscopies, and it is possible to use available microelectronics fabrication techniques to build such features without great difficulty.

We are very actively pursuing this work, and we hope that it may lead to important technological developments in catalyst design while at the same time providing a wealth of spatiotemporal patterns to analyze and study.

1.2 PUBLICATIONS

1.2.1 REFEREED JOURNALS

PUBLISHED

1. Hudson, J. L., M. Cube, R. A. Adomaitis, I. G. Kevrekidis, A. S. Lapedes, and R. M. Farber, "Nonlinear Signal Processing and System Identification: Applications to Time Series from Electrochemical Reactions," Chem. Eng. Sci. 45, 2075-2081 (1990).

2. Rico-Martinez, R., K. Krischer, I.G. Kevrekidis, M. Kube, and J. L. Hudson, "Discrete- vs Continuous-Time Nonlinear Signal Processing of Cu Electrodissolution Data," Chem. Eng. Comm. 118, 25-48 (1992).

3. Krischer, K., R. Rico-Martinez, I. G. Kevrekidis, H. H. Rotermund, G. Ertl, and J. L. Hudson, "A Heterogeneously Catalyzed Reaction with Spatiotemporal Variations: Model Identification Using Nonlinear Signal Processing," AIChEJ 39, 89-98 (1993).

4. Kube, M. C., S. T. Pride, and J. L. Hudson, "Local Analysis of Time Series from the Oscillatory Electrocatalytic Reduction of Hydrogen Peroxide," Chaos, Solitons and Fractals 3, 495-507 (1993).

5. Hudson, J. L., J. Tabora, K. Krischer, and I. G. Kevrekidis, "Spatiotemporal Period Doubling during the Electrodissolution of Iron," Phys. Letters A 179, 355-363 (1993).

6. Kevrekidis, I. G., Rico-Martinez, M., Ecke, R. E., Farber, R. M. and Lapedes, A. S., "Global Bifurcations in Rayleigh Benard Convection: experiments, empirical maps and numerical bifurcation analysis", Physica D, 51, pp.342-362 (1994)

7. Graham, M. D., Kevrekidis, I. G., Asakura, K., Krischer, K., Rotermund, H.-H. and Ertl, G., "Effects of boundaries on pattern formation: catalytic oxidation of CO on Pt", Science, 264, pp.80-82 (1994)

8. Pride, S. T., J. R. Scully, and J. L. Hudson, "Metastable Pitting of Aluminum and Criteria for the Transition to Stable Pit Growth," *J. Electrochem. Soc.* **141**, 3028-3040 (1994).

9. Hudson, J. L., "Chaos during heterogeneous chemical reactions", in the Proceedings of the 2nd Experimental Chaos Conference, W. Ditto, S. Vohra, M. Shlesinger and M. Spano, eds., World Scientific (1995)

10. Peckham, B. B., C. Frouzakis and I. G. Kevrekidis: "Bananas and Banana Splits: A Parametric Degeneracy in the Hopf Bifurcation for Maps", *SIAM J. Math. Anal.*, 26, pp. 190-217, (1995)

11. Letellier, C., L. Le Sceller, P. Dutertre, G. Gouesbet, Z. Fei, and J. L. Hudson, "Topological characterization and global vector field reconstruction of an experimental electrochemical system," *J. Phys. Chem.* (1995).

IN PRESS OR SUBMITTED

12. Graham, M. D., I. G. Kevrekidis, J. L. Hudson, G. Veser, K. Krischer, and R. Imbihl, "Dynamics of concentration Patterns of the NO+CO reaction on Pt: Analysis with the Karhunen-Loeve Decomposition," in press, Chaos, Solitions and Fractals (1995).

13. Graham, M. D. and Kevrekidis, I. G. "Alternative approaches to the Karhunen Loeve decomposition for model reduction and data analysis", Computers and Chemical Engineering, in press, (1995)

14. Graham, M. D., Kevrekidis, I. G., Baer, M., Asakura, K., Rotermund, H.-H. and Ertl, G., "Catalysis on microstructured Surfaces", in press, Phys. Rev. E. (1995)

15. Glasser, B., I. G. Kevrekidis and S. Sundaresan, "One- and Two-Dimensional Traveling Wave Solutions in Gas-Fluidized Beds", submitted to *J. Fluid Mech.*, February 1995, with B. Glasser and S. Sundaresan.

16. Haas, G., M. Baer, I. G. Kevrekidis, P. Rasmussen, H.-H. Rotermund and G. Ertl, "Observations of front bifurcations in controlled geometries: from one to two dimensions", to *Phys. Rev. Letters*, January 1995,

17. Sayer, Jon. C. and J. L. Hudson, "Spatiotemporal Patterns on a Ring Electrode," I&EC Research (1995).

1.2.2 PROCEEDINGS

18. R. Adomaitis, C. Frouzakis, I. G. Kevrekidis, M. Golden and B. E. Ydstie, "The structure of basin boundaries in a simple adaptive control system", in Chaotic Dynamics, Theory and Practice, Proceedings of the NATO 1992 advanced summer institute, Patras, summer 1992, T. Bountis, editor, pp.195-210, Plenum, NY (1992).

19. I. G. Kevrekidis, R. A. Adomaitis and C. E. Frouzakis, "Global Stability Analysis of an Adaptively Controlled Mixing Tank Experiment" Proceedings of the 1992 American Control Conference/WP4 pp.1039-1043 Chicago, 1992,

20. R. Rico-Martinez, I. G. Kevrekidis and R. A. Adomaitis, "Noninvertibility in Neural Networks", Proceedings of the 1993 IEEE Conference in Neural Networks, San Francisco, pp.382-386 (Refereed)

21. R. Rico-Martinez and I. G. Kevrekidis, "Continuous time modeling of nonlinear systems: a neural-network based approach", Proceedings of the 1993 IEEE Conference in Neural Networks, San Francisco, pp.1522-1525 (Refereed)

22. R. M. Farber, A. Lapedes, R. Rico-Martinez and I. G. Kevrekidis, "Identification of Continuous-Time Dynamical Systems: Neural Network Based Algorithms and Parallel Implementation", in Proceedings of the 6th SIAM Conference on Parallel Processing for Scientific Computing, R. Sincovec et al., eds., pp.287-291, SIAM Publications, Philadelphia (1993).

23. Rico-Martinez, R., Kevrekidis, I. G. and R. A. Adomaitis, "Nonivertible Dynamics in neural network models, Proc. 28th IEEE Annual Conference on Information Sciences and Systems, Princeton, N.J. (March 1994)

24. Rico-Martinez, R., Anderson, J. S. and I. G. Kevrekidis, "Continuous-time nonlinear signal processing: a neural network based approach for gray box identification", in the Proceedings of the 1994 IEEE Workshop on Neural Networks for Signal Processing, Ermioni, Greece (September 1994) (refereed)

1.2.3 CHAPTERS IN BOOKS:

25. Hudson, J. L., "Chaos in Electrochemical Systems," in Chaos in Chemistry and Biochemistry, R. J. Field and L. Gyorgyi, eds., World Scientific (1993).

26. Scully, J. R., S. T. Pride, and J. L. Hudson, "Analysis of Electrochemical Noise from Metastable Pitting in Al, aged Al- 2%Cu, and AA 2024-T3," Electrochemical Noise Measurement for Corrosion Applications ASTM (1995).

27. Rico-Martinez, R., Kevrekidis, I. G. and Krischer, K., "Nonlinear System Identification using Neural Networks: Dynamics and Instabilities" invited chapter to appear in "Neural Networks for Chemical Engineers", A. Bulsari, ed., Elsevier, in press (1995)

1.2.4 Presentations:

- Plenary Lecture, 12th annual meeting of the Canadian Applied Mathematics Society, Otawa, May 1991 (IGK)
- Invited Talk, 3d annual Soviet-American Conference on Chaos, Woods Hole, MA, July 1991 (IGK)
- Invited Lecture, NATO Advanced Research Workshop on Homoclinic Chaos, Brussels, Summer 1991 (JLH)
- NATO Advanced Summer Institute, Patras, Greece, July 1991
- AIChE annual meeting, November 1991
- Invited Talk, Model Reduction for Bifurcation Calculations, 8th U.C. Conference (IGK) of Nonlinear Science, U.C. Irvine, February 1992
- Research in Progress Symposium, NACE Meeting, April, 1992.
- 1992 ACC, Chicago
- Plenary Lecture Some nonlinear dynamic features of adaptive control systems, 1st World Congress of Nonlinear Analysts, Tampa, Fla. August 1992 (IGK)

- 2nd International Symposium on Corrosion of Electronic Materials and Devices, ECS, Toronto, October, 1992.
- Invited Address Noninvertibility and the Dynamics of Adaptive Control Systems", SIAM Conference on Control, Minneapolis, September 1992 (IGK)
- AIChE annual meeting, November, 1992. (three presentations)
- AIChE annual meeting, November, 1993.
- 6th SIAM Conference on Parallel Processing for Scientific Computing, Norfolk, VA, March 1993
- Invited Talk, Workshop on Computational Methods for Nonlinear Phenomena, Mathematisches Forschungsinstitut Oberwolfach, Oberwofach, Germany, January 1993 (IGK)
- Invited Talk, CHAMPP Workshop on Alternatives to General Circulation Models, organized by LLNL/DOE, Berkeley, CA, February 1993 (IGK)
- Invited Talk, Workshop on Applications of Pattern Formation, The Fields Institute of Research in the Mathematical Sciences, Waterloo, Ontario, Canada, March 1993, (IGK)
- 1993 IEEE International Conference on Neural Networks, San Francisco (two presentations)
- 13th General Europhysics Conference, Regensburg, March 1993
- Invited Talk, Workshop on Low-Dimensional Approximations of Nonlinear Dynamical Systems, Army High Performance Computing Research Center, University of Minnesota, Minneapolis, May 13-15 1993 (IGK)
- 1993 American Control Conference, San Francisco, June 1993 .
- IEEE Mediterranean Control Conference, Chania, Greece, June 1993 (two presentations)

- SIAM Annual Meeting, Philadelphia, PA, July 1993
- Invited·Talk, Hereaus Foundation Workshop on Pattern Formation in Distributed Systems, Potsdam, Germany, September 1993 (IGK)
- Invited Lecture,2nd Experimental Chaos Conference," Arlington, Virginia, Fall 1993 (JLH)
- AIChE annual meeting, November, 1994. (three presentations)
- Dynamics Days 94, Duke University, Durham, NC, January 1994 (presentation & poster)
- 28th IEEE conference on Information Sciences and Systems, March 1994
- SIAM annual meeting, San Diego, July 1994
- Invited Talk Gordon Conference on Chemical Oscillations, Rhode Island, August 1994 (IGK)
- Short presentation - Poster, 1994 IEEE Workshop on neural networks for signal processing, Ermioni, Greece
- AIChE annual meeting, November 1994 (three presentations)
- Invited Lecture, Symposium on Mathematical Modeling of Electrochemical Systems, AIChE. (JLH)
- Workshop on Noninvertible Dynamical Systems, NSF Geometry center, University of Minnesota, March 1995 (three presentations).

There were also approximately fifty Departmental and Industrial seminars about this work over the last three-four years, approximately ten of them given in German Universities by Prof. Hudson during his visit as a senior Humboldt Fellow.

1.3 HONORS, AWARDS, PRIZES :

John L. Hudson was awarded the R. H. Wilhelm Award of the AIChE for his work in Chemical Reaction Engineering; he also was named Honorary Professor, East China University for Science and Technology, Shanghai, China. He delivered the McCabe Lecture at North Carolina State University in 1993. He also is a Senior Humboldt Fellow.

Yannis G. Kevrekidis was promoted to Full Professor in the Department of Chemical Engineering in 1994 and became Senior Faculty in the Program in Applied and Computational Mathematics in 1993; he was awarded the Allan P. Colburn award of the American Institute of Chemical Engineers in 1994, given to a younger member of the Institute; delivered the Allan P. Colburn lecture at the U. of Delaware in 1994, and was invited to teach a minicourse at the Istituto Tecnologico de Celaya in Mexico (Gto) 1995. He was also selected as the Stanislaw Ulam Visiting Scholar at the Center for Nonlinear Studies / Theoretical Division of Los Alamos National Laboratory, Los Alamos, NM, for the academic year 1994-95, and won (along with two collaborators) the SIAM best video/poster presentation prize in the 1994 SIAM annual meeting.